Geometrical Analysis of a Bacterial Cellulose-Based Sensing Element

Giovanna Di Pasquale[1], Salvatore Graziani[2], Antonio Licciulli[3], Rossella Nisi[4], Antonino Pollicino[5],Carlo Trigona[2]

[1]D.S.C., Dipartimento di Scienze Chimiche
University of Catania, Viale Andrea Doria 6,
95125, Catania, Italy
[2]D.I.E.E.I., Dipartimento di Ingegneria Elettrica Elettronica e
Informatica, University of Catania, Viale Andrea Doria 6,
95125, Catania, Italy
[3]Antonio Licciulli, Dipartimento di Ingegneria dell'Innovazione,
University of Lecce, Via per Arnesano
73100, Lecce, Italy
[4]Rossella Nisi, BioFaber
via Luigi di Savoia 19
Mesagne, Brindisi, Italy
[5]D.I.C.A.R., Dipartimento di Ingegneria Civile e Architettura
University of Catania, Viale Andrea Doria 6,
95125, Catania, Italy
salvatore.graziani@dieei.unict.it, carlo.trigona@dieei.unict.it

***Abstract*—With the advent of IoT, industry 4.0 and precision agriculture an increasing interest has been devoted to sensors and transducers based on new materials and eco-friendly technologies. Recently, Bacterial Cellulose (BC) has been investigated as an intriguing solution for the realization of green devices and electronics. BC has excellent mechanical properties, it is fully biodegradable and greener then classical plant-derived cellulose. In this paper we investigate the influence of the geometry of BC based compounds on their mechano-eletrical transduction capability. An experimental campaign has been addressed obtaining very intriguing results.**

Keywords—bacterial cellulose, ionic liquids, greener transducers, biodegradable sensor, geometry analysis

I. INTRODUCTION

The society is undergoing deep changes, which pose technological challenges and require for the development of new production strategies. The aging of the population is one of the most relevant trends in western society. Distributed sensing system can assure the continuous monitoring of the elderly, both for assuring their wellness and prolong their active life and detection performance [1-3]. The massive distribution of autonomous sensing system, on one hand, will require for the development of low-cost technologies, on the other, will promote a tendency of using such systems as consumables [4,5]. Such an attitude raises concerns about the constraints that new sensing systems need to satisfy and on their after-life fate. A large fraction of envisaged sensing systems is required to be flexible, wearable, or even biocompatible [6-8]. In addition, taking into account their pervasive diffusion, we need production strategies that respect the environment [9,10]. Last, but not less important, since such systems will be low-cost and disposable, recyclable and/or biodegradable devices are of interest [10,11].

Paper has been widely investigated, in the past years, like a suitable candidate for next-generation electronics technology [12]. It has excellent mechanical properties and is fully biodegradable [13]. Eventually, it can be processed for realizing electronics. Notwithstanding such tantalizing properties, cellulose is commonly derived from vegetal sources. The pulp industry processes wood as raw material for papermaking [14]. Unfortunately, the involved industrial process requires significant amounts of energy and resources (such as fresh water), which limit the attractiveness of plant-derived cellulose as a green based material.

Recently, Bacterial Cellulose (BC) has been investigated as an alternative candidate for the realization of green electronics [15]. Though BC has the same chemical structure as plant-derived cellulose, it is directly synthesized by some bacteria of the genera *Acetobacter*, *Sarcinaventriculi*, and *Agrobacterium*. Eventually, processes have been developed that allow for obtaining BC cellulose in laboratory condition in a kind of bio-industry. Compared to plant-derived cellulose BC is obtained with a greener production process, which works in typical laboratory conditions. The obtained BC is much purer than conventional cellulose.

Results have been proposed demonstrating the possibility of using compounds realized by using BC, covered by conductors, and infused by Ionic Liquids (ILs) for realizing electro-mechanic transduction [15-17]. Based on those studies, the authors have recently demonstrated that such BC based compounds can work also as mechano-electric transducers [18,19]. Here, as further development, the influence of the geometry of the device on its transduction capability is investigated.

More specifically, the investigated devices are realized in the form of a multilayer structure, where the BC is impregnated by ILs. Then two thin layers of Conducting Polymers (CPs) are deposited as the electrodes. Finally, samples with a rectangular shape are extracted and mounted in a cantilever configuration. When the composite is deflected, it produces an electrical reaction. More specifically, if it is connected in the open-circuit condition a voltage signal is produced.

The thickness of the investigated samples is fixed by the BC production process, since BC has been obtained by BioFaber in the form of sheets. The length has been, therefore, changed and the corresponding changes in the mechano-electrical (open voltage signal) performance have been investigated.

The paper is organized as follows: Section II will describe materials, the realized prototype, and the methods used to characterize the composite and its transducing properties. Section III reports and discuss the obtained results, while the concluding remarks are given in Section IV.

II. MATERIALS, PROTOTYPE AND METHODS

Materials

Black tea bags (Sir Bolton Company), sucrose (commercial product), Vinegar (commercial product), and Kombucha tea were purchased from the local shops. Sodium hydroxide (NaOH), and absolute ethanol (99.8%), were purchased from Sigma-Aldrich and used without further modification. Agar powder was purchased from A.C.E.F. and LB broth Miller was purchased from VWR. Escherichia coli culture BL21(DE3) strain was purchased from Merck KGaA, Germany. PEDOT/PSS, was purchased as an aqueous dispersion, CLEVIOS PHCV4, by H.C.Starck. EMIM-BF4 was purchased by Alfa Aeser.

Preparation of BC base compounds

BC pellicles were obtained from the Kombucha strains, which is produced by the fermentation process of the sweetened black tea with Acetobacter strains. The tea fungus that is composed of upper cellulosic pellicle and a lower liquid broth was activated every two weeks, according to the procedure described by Chen and Liu with some modifications [20]. The culture medium was prepared by adding 70 g of sucrose and two tea bags (black tea, ~ 4.00 g) to the boiling water (1 l). Then, the mixture was left to steep for 15 min and, after removing the tea bags, the pH value of the broth was adjusted to 2.7–3.0, by adding 10 ml of acetic acid for each liter of the broth. The addition of acetic acid at the beginning of the fermentation process prevents the formation of molds and protects against undesirable microorganisms. Finally, the cellulosic pellicle pieces (3% w/v) and liquid broth (10% v/v) of the tea fungus were added to the cooled tea broth.

A Teflon stopper was attached to the lower part of the beaker to block the pellicle. The fermentation process was carried out at room temperature (28 °C) for 15 d in a static culture condition. In this period, new pellicles of cellulose were grown on the surface of the broth. These pellicles and the tea fungus were used to inoculate new fermentations. Then, the produced pellicles or named as nanofibrillated BC were thoroughly washed with distilled water and boiled in 0.5 M NaOH solution for 2 h, to remove any impurities and the attached cells, and finally washed with deionized water and stored in wet condition. In order to extract the endotoxin from the BC samples, they were placed in a depyrogenated sample container and boiled with endotoxin-free water 4 times.

Then BC was dried in an oven for 4 h at 70 °C and freeze-dried for 1 d, in order to eliminate the water content. Samples of the BC were cut, infused by the EMIM-BF4, as the ionic liquid, and covered by the PEDOT-PSS, as flexible the electrodes.

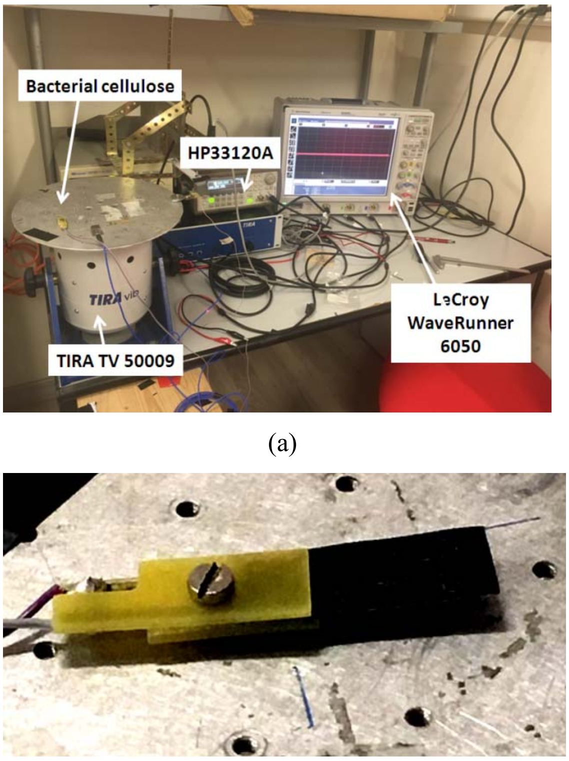
Characterization methods

In order to analyze the sensor as a function of its geometry, a suitable experimental setup has been conceived.

In particular, Fig.1(a) shows the entire architecture which is composed of:

- a shaker (TIRA TV 50009), to excite the structure.
- A signal generators HP33120A, to impress a sinusoidal waveform.
- A digital Teledyne LeCroyWaveRunner 6050, to acquire the signals.

Fig. 1(b) shows a sample of the BC-based compound mounted between two rigid electrodes, used for collecting the generated electrical signal.



(a)

(b)

Fig.1. *(a) Experimental setup, (b) the BC with the two electrical contacts on the top and on the bottom.*

III. EXPERIMENTAL RESULTS AND DISCUSSION

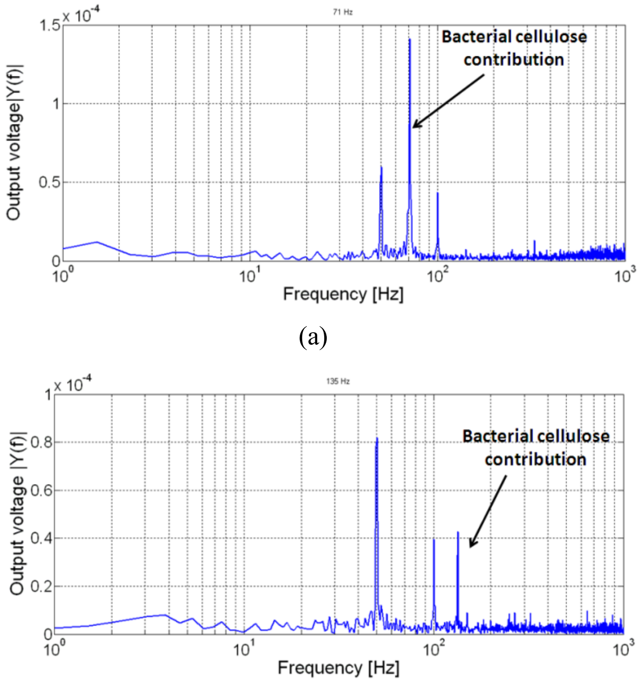
This section is devoted to studying the effect of the geometry of a BC based transducer on its output, at the mechanical resonant frequency.

The investigation has been conducted considering samples with a width of 11 mm and a thickness of about 0.25 mm. Devices, whose lengths were 22 mm, 17 mm, 12 mm, and 7 mm were investigated. An analysis of the output voltage as a function of the impressed acceleration, when the transducer geometry is changed, has been performed in the frequency domain.

The measurements have been conducted by forcing a sinusoidal signal through the signal generator (see Section II) at the mechanical resonant frequency of each device. It is worth noting that for the four case studies, the following mechanical resonant frequency values have been experimentally estimated: 71 Hz (device #1 length of 22 mm), 110 Hz (device #2 length of 17 mm), 133 Hz (device #3 length of 11 mm) and 135 Hz (device #4 length of 7 mm).

More details concerning the spectrum of the output voltage can be shown in Fig.2(a) and Fig.2(b), where the FFT of the output for devices #1 and #4 are presented, respectively.

Fig.3 shows the normalized output voltage as function of the normalized imposed acceleration. The voltage of each sample has been normalized to the output voltage of the first sample, i.e. the device having a length of 22 mm.



(a)

(b)

Fig.2. *FFT of the output voltage of (a) BC having a length of 22 mm, (b) BC having a length of 7 mm. Two other spikes can be seen in the figure, which refer to the powering line (50 Hz and 100 Hz contributions, respectively).*

Furthermore, the acceleration values have been normalized to the acceleration a1, which refers to the first sample. The value a2 is the acceleration level at frequency 110 Hz, normalized to a1, and so on for a3 (at 133 Hz) and a4 (at 135 Hz) respectively. Fig.4 shows the normalized output voltage as function of the normalized length of the beam. The normalization has been performed considering the length of the device #1.

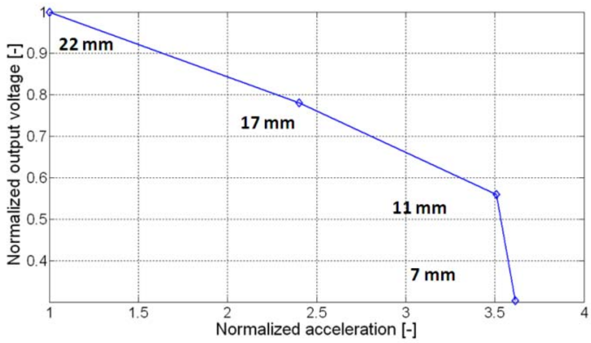


Fig.3. *Normalized output voltage as respect the normalized acceleration.*

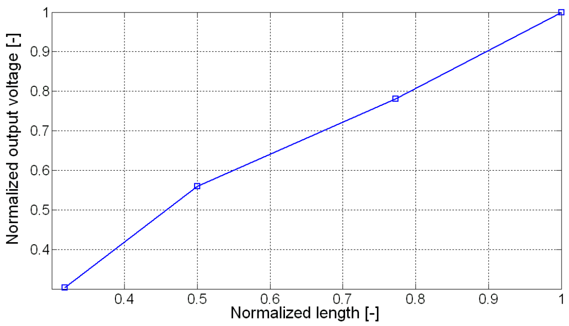


Fig.4. *Normalized output voltage as respect the normalized length.*

IV. CONCLUSIONS

In this paper the dependence of the transduction capabilities of a BC based compound, which can be used as sensing element, on its geometry has been accomplished. The conceived sensor regards a cantilever beam and it is based on a nanocomposite consisting of a green and biodegradable solution based on a BC, impregnated with ILs, and covered with a conductive polymer. The device is able to generate an output voltage in the presence of an impressed acceleration, which causes a deformation of the transducer. Several geometries have been investigated and the effect of a length reduction has been studied. Results demonstrate how the geometry variation influence the output signal. The work is in progress with an exhaustive characterization also taking into the account the effect of other physical properties on the performance of the device.

REFERENCES

[1] D.L. Carnì, D. Grimaldi, F. Lamonaca, L. Nigro, P.F. Sciammarella, "From distributed measurement systems to cyber-physical systems: A design approach", International Journal of Computing, vol. 16, pp. 66-73, 2017.

[2] F. Tramarin, S. Vitturi, M. Luvisotto, “IEEE 802.11 n for Distributed Measurement Systems”, 2017 IEEE International Instrumentation and Measurement Technology Conference (I2MTC), pp. 1-6, 2017.

[3] S. Troja, E. Egger, P. Francescon, A. Gueli, A. Kacperek, M. Coco, R. Musmeci, and A. Pedalino, “2d and 3d dose distribution determination in proton beam radiotherapy with gafchromic film detectors,” Technology and Health Care, vol. 8, no. 2, pp. 155–164, 2000.

[4] D.J. Inman, B.L. Grisso, “Towards autonomous sensing”, Smart Structures and Materials 2006: Sensors and Smart Structures Technologies for Civil, Mechanical, and Aerospace Systems, vol. 6174, pp. 61740T, 2006.

[5] B. Ando, S. Baglio, C. Trigona, “Autonomous sensors: From standard to advanced solutions”, IEEE Instrumentation & Measurement Magazine, vol. 13, pp. 33-37, 2010.

[6] X. Ding, H. Cao, X. Zhang, M. Li, Y. Liu, “Large Scale Triboelectric Nanogenerator and Self-Powered Flexible Sensor for Human Sleep Monitoring”, Sensors, vol. 18, pp. 1713, 2018.

[7] D. Spirjakin, A. Baranov, S. Akbari, “Wearable wireless sensor system with RF remote activation for gas monitoring applications”, IEEE Sensors Journal, vol. 18, pp. 2976-2982, 2018.

[8] P.J. Babu, A.M. Raichur, M. Doble, “Synthesis and characterization of biocompatible carbon-gold (C-Au) nanocomposites and their biomedical applications as an optical sensor for creatinine detection and cellular imaging”, SENSORS AND ACTUATORS B-CHEMICAL, vol. 258, pp. 1267-1278, 2018.

[9] S. Kanaparthi, “Eco-friendly engineered low-cost conformable capacitive sensing device to measure water content in adulterated ethanol fuel”, Flexible and Printed Electronics, vol. 3, pp 015009, 2018.

[10] S. Yamada, H. Toshiyoshi, “A Water Dissolvable Electrolyte with an Ionic Liquid for Eco-Friendly Electronics”. Small, vol. 14, pp. 1800937, 2018.

[11] A. Yuen, L.H. Porcarelli, R. Aguirresarobe, A. Sanchez-Sanchez, I. Del Agua, U. Ismailov, H. Sardon, “Biodegradable polycarbonate iongels for electrophysiology measurements”, Polymers, vol. 10, pp. 989, 2018.

[12] J.C. Cunningham, P.R. DeGregory, R.M. Crooks,x “New functionalities for paper-based sensors lead to simplified user operation, lower limits of detection, and new applications”, Annual Review of Analytical Chemistry, vol. 9, pp. 183-202, 2016.

[13] S. Kanaparthi, S. Badhulika, “Low cost, flexible and biodegradable touch sensor fabricated by solvent-free processing of graphite on cellulose paper”. Sensors and Actuators B: Chemical, vol. 242, pp. 857-864, 2017.

[14] R. Mangayil, S. Rajala, A. Pammo, E. Sarlin, J. Luo, V. Santala, M. Karp, S. Tuukkanen, “Engineering and Characterization of Bacterial Nanocellulose Films as Low Cost and Flexible Sensor Material”, ACS Applied Materials and Interfaces, vol. 9, pp. 19048-19056, 2017.

[15] J.H. Jeon, I.K. Oh, C.D. Kee, S.J. Kim, “Bacterial Cellulose actuator with electrically driven bending deformation in hydrated condition”, Sensors and Actuators B: Chemical, vol. 146, pp. 307-313, 2010.

[16] F. Esa, S.M. Tasirin, N.A. Rahman, “Overview of bacterial cellulose production and application”, Agriculture and Agricultural Science Procedia, vol. 2, pp. 113-119, 2014.

[17] S.S. Kim, J.H. Jeon, C.D. Kee, I.K. Oh, “Electro-active hybrid actuators based on freeze-dried bacterial cellulose and PEDOT:PSS”, Smart Materials and Structures, vol. 22, pp. 085026, 2013.

[18] G. Di Pasquale, S. Graziani, A. Pollicino, C. Trigona, ““Paper”Based Sensor for Deformation Measurements”, IEEE-I2MTC2019, in press.

[19] . Di Pasquale, S. Graziani, A. Pollicino, C. Trigona, “Green Inertial Sensors based on Bacterial Cellulose”, IEEE-SAS2019, in press.

[20] C. Chen, B.Y. Liu, “Changes in major components of tea fungus metabolites during prolonged fermentation”, Journal of applied microbiology, vol. 89, pp. 834-839, 2000.